

# Critical points of the optimal quantum control landscape: a propagator approach

Tak-San Ho · Herschel Rabitz · Gabriel Turinici

Received: date / Accepted: date

**Abstract** Numerical and experimental realizations of quantum control are closely connected to the properties of the mapping from the control to the unitary propagator [14]. For bilinear quantum control problems, no general results are available to fully determine when this mapping is singular or not. In this paper we give sufficient conditions, in terms of elements of the evolution semigroup, for a trajectory to be non-singular. We identify two lists of “way-points” that, when reached, ensure the non-singularity of the control trajectory. It is found that under appropriate hypotheses one of those lists does not depend on the values of the coupling operator matrix.

**Keywords** quantum control · singular control · landscape analysis in quantum control

**PACS** 32.80.Qk · 02.30.Yy

**Mathematics Subject Classification (2000)** 15A57 · 49J20 · 65Z05

## 1 Introduction

Manipulating the evolution of physical systems at the quantum level has been a longstanding goal from the very beginnings of laser technology. Investigations

---

T.-S. Ho  
Department of Chemistry, Princeton University, Princeton, NJ 08544, USA  
E-mail: tsho@princeton.edu

H. Rabitz  
Department of Chemistry, Princeton University, Princeton, NJ 08544, USA  
E-mail: hrabitz@princeton.edu

G. Turinici  
CEREMADE, Université Paris Dauphine, Place du Marechal de Lattre de Tassigny, 75016 Paris, France  
E-mail: Gabriel.Turinici@dauphine.fr

in this area accelerated after the introductions of (optimal) control theory tools [11], which greatly contributed to the first positive experimental results, see [3, 13, 16, 4, 5, 10, 12] and references herein.

Consider a quantum system in the presence of a control field  $\epsilon(t) \in \mathbb{R}$ ,  $t \geq 0$ . In the electric dipole approximation and in the density matrix formulation, the underlying time-dependent density matrix  $\rho(t)$  satisfies the Liouville-von Neumann equation

$$\begin{aligned} i \frac{\partial}{\partial t} \rho(t) &= [H_0 - \epsilon(t)\mu, \rho(t)] \\ \rho(t=0) &= \rho_0 \end{aligned} \quad (1)$$

where  $H_0$  is the field-free Hamiltonian (including the potential) and  $\mu$  is the dipole moment of the system. In the analysis, we will suppose that the system contains  $N$  levels, thus,  $H_0$ ,  $\mu$  and  $\rho(t)$  are all  $N \times N$  Hermitian matrices. In the following, for simplicity, we further assume that  $H_0$  and  $\mu$  are real and symmetric matrices. Moreover, to avoid trivial settings, the dipole moment operator  $\mu$  is assumed to have zero trace, i.e.,

$$\text{Tr}(\mu) = 0. \quad (2)$$

In the following we adopt the notation  $isu(N)$  to denote the set of all complex  $N \times N$  Hermitian matrices with zero trace. Recall that  $\rho(t) = U(t, 0)\rho(0)U^*(t, 0)$ , where the unitary propagator  $U(t, 0) \in U(N)$ ,  $t \geq 0$  is the solution of the time-dependent Schrödinger equation

$$\begin{aligned} i \frac{\partial}{\partial t} U(t, 0) &= (H_0 - \epsilon(t)\mu)U(t, 0) \\ U(t=0, 0) &= I_N. \end{aligned} \quad (3)$$

The control goal can be expressed in terms of a (self-adjoint) operator  $O$  in that the corresponding functional to be optimized (maximized) is  $\langle O \rangle(T) = \text{Tr}(\rho(T)O)$ . Accordingly, a maximum value of  $\langle O \rangle(T)$  indicates that the control is of optimal quality and *vice-versa*.

At the heart of a good understanding of any quantum control problem, including the efficiency, accuracy, and stability of quantum control experiments as well as simulations, lies essential features of the control landscape, depicting the functional dependence of the observable on the control field, i.e. the mapping:  $\epsilon(t) \mapsto \langle O \rangle(T)$ . The slopes (gradient) and the curvatures (Hessian) are key characteristics of the control landscape. In particular, the topologies at and around the critical points, in which the slopes vanish, can shed important light on the questions, for example, of what makes quantum control experiments (simulations) apparently “easy to perform” and why quantum control beats the “curse of dimensionality” (i.e., overcomes the anticipated exponentially growing effort required when searching over increasing numbers of control variables) [9]. A thorough study of the underlying control landscape should give insight into the classes of future feasible quantum control experiments, especially those involving complex molecules and materials.

## 2 Motivation: landscape analysis and beyond

The search procedures optimize the control quality  $\langle O \rangle(T)$  with respect to variations in the control  $\epsilon(t)$ . The mapping  $\epsilon(t) \mapsto \langle O \rangle(T)$  is the composition of two maps: a control map  $\epsilon(t) \mapsto U(T, 0)$  and a "kinematic" map  $U(T, 0) \mapsto \langle O \rangle(T)$ . A key issue is whether there exist critical points that are suboptimal solutions. To analyse the critical points it is expedient to consider these two maps separately.

Previous analyses [14] have investigated the mapping from the propagator to the observable i.e.  $U(T, 0) \mapsto \langle O \rangle(T)$ ; the critical points correspond to the equation

$$\frac{\delta \langle O \rangle(T)}{\delta U(T, 0)} = 0. \quad (4)$$

Assuming controllability hypothesis [2, 15], this equation is satisfied if and only if

$$[U^*(T, 0)OU(T, 0), \rho(0)] = 0, \quad (5)$$

i.e., we obtain a necessary and sufficient condition for the kinematic critical points [7]. The optimization "landscape", when looked upon in terms of this so called "kinematic" mapping, is therefore exempt of suboptimal critical points.

To extend the landscape analysis beyond the kinematic setting, i.e. to investigate the dependence on the control itself, we need to analyse the solutions of the following critical point condition [9]

$$\frac{\delta \langle O \rangle(T)}{\delta \epsilon(t)} = 0. \quad (6)$$

The solutions of this equation are among the critical points of the control map  $\epsilon(t) \mapsto U(T, 0)$ ; it was shown in [17] that this mapping is nonsingular when all matrix entries of the dipole moment

$$\hat{\mu}(t) = U^*(t, 0)\mu U(t, 0), \quad (7)$$

as functions of  $t \in [0, T]$ , are linearly independent (to the extent possible), or equivalently if the dipole moment matrices  $\hat{\mu}(t)$ 's span the space of all zero-traced Hermitian matrices  $isu(N)$ .

In previous works, it has always been assumed that "full controllability" implies "fully linear independence" of the dipole moment matrix entries. An assertion of the linear independence of dipole moment matrix entries is also essential for understanding the landscape of unitary transformation control problem [8]. In this paper, we present a rigorous analysis of this assertion and identify properties of the trajectory, in terms of the propagators, that are sufficient to ensure the linear independence of entries of  $\hat{\mu}(t)$ . In addition, the analysis of which controls correspond to a non-singular mapping  $\epsilon(t) \mapsto U(T, 0)$  or  $\epsilon(t) \mapsto \langle O \rangle(T)$  is also relevant, e.g., for stabilization purposes, error cancellation, maintaining coherence, etc.

### 3 Dipole dependent way-points

Denote by  $\mathbb{L}$  the Lie algebra generated by  $-iH_0$  and  $-i\mu$  as a sub-algebra of  $u(N)$ . We will suppose that the system is density matrix controllable, or equivalently

$$\mathbb{L} = su(N) \text{ or } \mathbb{L} = u(N). \quad (8)$$

Note that when the system is density matrix controllable, then it is also wave function controllable, cf [1,6].

**Theorem 1** *Under assumptions (2) and (8) there exists a set  $\{U_1, \dots, U_{2N^2-2N}\}$  of unitary propagators (that we will call the set of "way-points") such that if  $U(t, 0)$  visits all of them i.e.  $U(t_k, 0) = U_k$  for some times  $t_k$ ,  $k = 1, \dots, 2N^2 - 2N$  the components  $f_{ij}(t)$  of the matrix  $\hat{\mu}(t)$  in (7) are linearly independent as functions of time.*

*Consequently, under the hypotheses (2) and (8) for  $T$  sufficiently large a control field  $\epsilon(t)$ ,  $t \in [0, T]$  exists such that functions  $f_{ij}(t)$  are linearly independent (or equivalently such that  $\hat{\mu}(t)$  spans the set of all zero-traced Hermitian matrices).*

**Remark 1** *Note that here there is no information required on the structure of the matrices  $H_0$  or  $\mu$ .*

**Proof** Since the Hermitian matrix  $\mu$  has zero trace it cannot be a constant and thus it has at least two different eigenvalues  $\lambda_1$  and  $\lambda_2$ .

Let us begin by some general conventions: for any  $i \neq j$  and any matrix  $M$  we denote by  $M_{<i,j>}$  its  $2 \times 2$  sub-matrix formed by the  $i$ -th and  $j$ -th rows and columns:

$$M_{<i,j>} = \begin{pmatrix} M_{ii} & M_{ij} \\ M_{ji} & M_{jj} \end{pmatrix}. \quad (9)$$

Moreover, when we alter the  $N \times N$  identity matrix by putting in the  $i$ -th and  $j$ -th rows and columns a given  $2 \times 2$  matrix  $D$ , we will denote the resultant  $N \times N$  matrix by  $D^{<i,j>}$  :

$$D^{<i,j>} = \begin{pmatrix} & & i & j & \\ & & & & \\ & & 1 & 0 & 0 & 0 & 0 \\ & & 0 & 1 & 0 & 0 & 0 \\ & & 0 & 0 & D_{11} & D_{12} & 0 \\ & & 0 & 0 & D_{21} & D_{22} & 0 \\ & & 0 & 0 & 0 & 0 & 1 \end{pmatrix} \begin{matrix} \\ \\ i \\ j \\ \\ \end{matrix} \quad (10)$$

We choose  $U_k$  in the following way: let  $l = 1, \dots, \frac{N(N-1)}{2}$  index the couples  $(i, j)$ ,  $i, j = 1, \dots, N$  with  $i < j$ .

-  $U_{4l+1}$  is such that  $\left(\hat{\mu}(t_{4l+1})\right)_{<i,j>} = \begin{pmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{pmatrix}$ ;

-  $U_{4l+2} = U_{4l+1} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}^{<i,j>}$ . Then  $(\hat{\mu}(t_{4l+2}))_{<i,j>} = \begin{pmatrix} \lambda_2 & 0 \\ 0 & \lambda_1 \end{pmatrix}$  and all other entries are identical to those of  $\hat{\mu}(t_{4l+1})$ .

-  $U_{4l+3} = \frac{1}{2}U_{4l+1} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}^{<i,j>}$ . Then

$$(\hat{\mu}(t_{4l+3}))_{<i,j>} = \frac{1}{2} \begin{pmatrix} \lambda_1 + \lambda_2 & \lambda_1 - \lambda_2 \\ \lambda_1 - \lambda_2 & \lambda_1 + \lambda_2 \end{pmatrix} \quad (11)$$

and all other entries are identical to those of  $\hat{\mu}(t_{4l+1})$ .

-  $U_{4l+4} = \frac{1}{2}U_{4l+1} \begin{pmatrix} i & 1 \\ 1 & i \end{pmatrix}^{<i,j>}$ . Then

$$(\hat{\mu}(t_{4l+4}))_{<i,j>} = \frac{1}{2} \begin{pmatrix} \lambda_1 + \lambda_2 & (\lambda_2 - \lambda_1)i \\ (\lambda_1 - \lambda_2)i & \lambda_1 + \lambda_2 \end{pmatrix} \quad (12)$$

and all other entries are identical to those of  $\hat{\mu}(t_{4l+1})$ .

Note that all  $\hat{\mu}(t_k) = U_k^* \mu U_k$  are Hermitian (and of zero trace). Let us take a zero trace Hermitian matrix  $Z$  such that

$$\langle Z, \hat{\mu}(t_k) \rangle = 0, \quad \forall k = 1, \dots, 4N. \quad (13)$$

where we used the canonical scalar product of Hermitian matrices  $\langle A, B \rangle = \text{Tr}(A^* B) = \text{Tr}(AB)$ . We recall also the definition of the norm induced by this scalar product  $\|A\| = \sqrt{\langle A, A \rangle}$ .

We denote  $\begin{pmatrix} x & y \\ y^* & z \end{pmatrix} = Z_{<i,j>}$ ; since  $\langle Z, \hat{\mu}(t_k) \rangle = 0$  for  $k = 4l + 1, \dots, 4l + 4$  it follows that  $\langle Z_{<i,j>}, (\hat{\mu}(t_k))_{<i,j>} \rangle$  is the same for  $k = 4l + 1, \dots, 4l + 4$ . The consequence is that  $\lambda_1 x + \lambda_2 z = \lambda_2 x + \lambda_1 z$  which together with  $\lambda_1 \neq \lambda_2$  implies  $x = z$ . Moreover we also have  $\lambda_1 x + \lambda_2 x = \frac{1}{2} \left( 2(\lambda_1 + \lambda_2)x + (\lambda_1 - \lambda_2)(y + y^*) \right)$  thus  $y + y^* = 0$ . Additionally, we have  $\lambda_1 x + \lambda_2 x = \frac{1}{2} \left( 2(\lambda_1 + \lambda_2)x + (\lambda_1 - \lambda_2)i(y^* - y) \right)$ , thus  $y = y^*$ . From  $y + y^* = 0$  and  $y = y^*$  we infer that  $y = 0$  and obtain  $Z_{<i,j>} = xI_2$  ( $I_2$  is the  $2 \times 2$  unit matrix). Since this is true for arbitrary  $i < j$  we obtain  $Z = xI_N$ . But, since  $\text{Tr}(Z) = 0$  it follows  $Z = 0$ , q.e.d.

For the second part of the conclusion we use the fact that under the hypotheses (2) and (8) the system is controllable thus for  $T$  sufficiently large a control field  $\epsilon(t)$ ,  $t \in [0, T]$  exists such that functions  $f_{ij}(t)$  are linearly independent.

#### 4 Dipole independent way-points

The result of the previous section can be interpreted as follows: as long as the control field ensures that the propagator will visit some specific unitary transformations, then the required linear independence of the time-dependent

elements of  $\hat{\mu}(t)$  is satisfied. Note that this set of unitary transformations (i.e., propagators) explicitly depends on the dipole moment. In the laboratory, procedures have been designed that are able to control the quantum evolution even in the absence of precise information on the dipole [11]. It is therefore possible in principle to experimentally find a control passing through a specified list of propagators when this list does not depend on the dipole entries. A universal procedure can therefore be implemented that will ensure the non-singularity of the mapping  $\epsilon(t) \mapsto U(T, 0)$  even when the dipole is unknown. Beyond the question of the landscape analysis, such a procedure can be useful in additional circumstances when the mapping is required to be non-singular, e.g. for stabilization purposes, etc. We will therefore investigate the following circumstance: suppose that the system is controllable and thus one can experimentally implement arbitrary propagators  $U \in SU(N)$ . Can we find a list of propagators (as in the previous section) which are independent of the precise values of entries of  $\mu$ ?

**Theorem 2** *Under assumptions (2) and (8) there exists a set of "way-points"  $\mathbb{W} \subset SU(N)$  independent of  $\mu$  such that if  $U(t, 0)$  visits all propagators in  $\mathbb{W}$  (i.e. for all  $U_k \in \mathbb{W}$  there exists  $t_k$  with  $U(t_k, 0) = U_k$ ) the components  $f_{ij}(t)$  of the matrix  $\hat{\mu}(t)$  in (7) are linearly independent as functions of time.*

**Proof** We denote for any  $U \in SU(N)$ :

$$\mathcal{C}_U = \{(Z, \mu) \in (isu(N))^2 \mid \|Z\| = \|\mu\| = 1, \text{Tr}(ZU^*\mu U) \neq 0\}. \quad (14)$$

This set is open because its complement is the solution of a linear equation in the entries of the matrices  $Z$  and  $\mu$ .

We prove now that for any  $Z, \mu \in isu(N)$  of unit norm there exists a  $U \in SU(N)$  such that  $\text{Tr}(ZU^*\mu U) \neq 0$ . To see this, diagonalize  $Z$  and  $\mu$ :  $Z = U_1^* D_1 U_1$  and  $\mu = U_2^* D_2 U_2$  with  $U_1, U_2$  unitary and  $D_1, D_2$  diagonal; we will denote by  $d_k^a$  the  $k$ -th diagonal entry of  $D^a$ . Recall that since  $Z, \mu$  are zero-traced  $\sum_{k=1}^N d_k^a = 0$  for  $a = 1, 2$ . Therefore, in each set  $(d_k^a)_{k=1}^N$  there are some elements that are strictly negative and some strictly positive. We will also suppose that the diagonalization is done in such a way that the diagonal elements are ordered from the lowest (that is strictly negative) to the highest, which is strictly positive. Then,

$$\text{Tr}(ZU^*\mu U) = \text{Tr}(U_1^* D_1 U_1 U^* U_2^* D_2 U_2 U) = \text{Tr}(D_1 V^* D_2 V) \quad (15)$$

with  $V = U_2 U U_1^* \in SU(N)$ . Take now  $U$  such that  $V$  is a permutation matrix corresponding to permutation  $\sigma$ . Then

$$\text{Tr}(D_1 V^* D_2 V) = \sum_{k=1}^N d_k^1 d_{\sigma(k)}^2. \quad (16)$$

If  $\sum_{k=1}^N d_k^1 d_{\sigma(k)}^2 = 0$  for any permutation  $\sigma$  then it follows in particular that  $\sum_{k=1}^N d_k^1 d_k^2 = \sum_{k=1}^N d_k^1 d_{N-k}^2$ , but the first member is always superior to the

second with the equality implying that one of the vectors  $d_k^a$  is constant which leads to a contradiction with the zero-trace hypothesis.

We therefore obtain that  $\cup_{U \in SU(N)} \mathcal{C}_U$  is a covering of the compact set  $\{(Z, \mu) \in (isu(N))^2 \mid \|Z\| = \|\mu\| = 1\}$  and thus one can extract a finite covering i.e. a finite set  $\mathbb{W}$  such that

$$\cup_{U \in \mathbb{W}} \mathcal{C}_U = \{(Z, \mu) \in (isu(N))^2 \mid \|Z\| = \|\mu\| = 1\}. \quad (17)$$

This means that for any  $Z$  and  $\mu$  in  $isu(N)$  (note that there is no need to impose the norm condition) there exists  $U \in \mathbb{W}$  such that  $Tr(ZU^*\mu U) \neq 0$ . Expressed otherwise, for any  $\mu \in isu(N)$  there does not exist  $Z \in isu(N)$  such that  $Tr(ZU^*\mu U) = 0$  for any  $U \in \mathbb{W}$ . This implies that  $\{U^*\mu U \mid U \in \mathbb{W}\}$  form an independent set of vectors in  $isu(N)$ , q.e.d.  $\square$

**Remark 2** *Theorem 2 only assures that such a set  $\mathbb{W}$  exists but does not give yet precise information on its size (which can be large) nor does it provide a constructive approach to identify in practice  $\mathbb{W}$ . The following result addresses these questions upon imposing an additional constraint on the dipole  $\mu$ .*

**Theorem 3** *Under assumptions (2) and (8) and*

$$\mu_{ij} \neq 0, \forall i \neq j. \quad (18)$$

*there exists a set of "way-points"  $\mathbb{S} \subset SU(N)$  such that if  $U(t, 0)$  visits all propagators in  $\mathbb{S}$  (i.e. for all  $U_k \in \mathbb{S}$  there exists  $t_k$  with  $U(t_k, 0) = U_k$ ) the components  $f_{ij}(t)$  of the matrix  $\hat{\mu}(t)$  in (7) are linearly independent as functions of time. Moreover the set  $\mathbb{S}$  can be chosen to be the same for all coupling operators  $\mu$  satisfying the hypotheses (2), (8), (18).*

**Proof** Suppose that the entries of  $\hat{\mu}(t)$  are not linearly independent i.e. a matrix  $Z$ ,  $Tr(Z) = 0$  exists such that  $Z$  is orthogonal to all matrices  $U^*\mu U$ ,  $\forall U \in \mathbb{S}$ , i.e.,  $Tr(ZU^*\mu U) = 0 \forall U \in \mathbb{S}$ .

Let us compute  $U^*\mu U$ : for  $\mu = \begin{pmatrix} \mu_{11} & \mu_{12} \\ \mu_{21} & \mu_{22} \end{pmatrix}$  (here  $\mu_{11}$  is a  $2 \times 2$  matrix,  $\mu_{22}$  a  $N - 2 \times N - 2$  matrix, etc.) and  $U = W^{<1,2>} = \begin{pmatrix} W & 0 \\ 0 & I_{N-2} \end{pmatrix}$

$$U^*\mu U = \begin{pmatrix} W^*\mu_{11}W & W^*\mu_{12} \\ \mu_{21}W & \mu_{22} \end{pmatrix}. \quad (19)$$

A typical example of  $W$  is  $\begin{pmatrix} \cos(\phi) & \sin(\phi) \\ \sin(\phi) & -\cos(\phi) \end{pmatrix}$ . The entries of  $W^*\mu_{11}W$  will contain terms of second order in  $\cos(\phi)$  and  $\sin(\phi)$ :  $\cos^2(\phi) = \frac{\cos(2\phi)+1}{2}$  and  $\sin^2(\phi) = \frac{1-\cos(2\phi)}{2}$ ,  $\sin(\phi)\cos(\phi) = \frac{\sin(2\phi)}{2}$ ; the entries of  $W^*\mu_{12}$  and  $\mu_{21}W$  will contain only first order terms in  $\cos(\phi)$  and  $\sin(\phi)$  while  $\mu_{22}$  does not depend on  $\phi$  at all.

We recall that for any  $\theta \in \mathbb{R}$  the functions  $1, \cos(\theta), \sin(\theta), \cos(2\theta), \sin(2\theta)$  are linearly independent, which will be used in the following fashion

**Lemma 1** *There exist five real values of  $\theta$ , for instance  $\Theta = \{0, \pi/3, \pi/2, \pi, 3\pi/3\}$  such that*

$$\begin{aligned} &\alpha_1 + \alpha_2 \cos(\theta) + \alpha_3 \sin(\theta) \\ &+ \alpha_4 \cos(2\theta) + \alpha_5 \sin(2\theta) = 0 \quad \forall \theta \in \Theta \end{aligned} \quad (20)$$

*implies*

$$\alpha_1 = \dots = \alpha_5 = 0. \quad (21)$$

We now introduce into the set  $\mathbb{S}$  the matrices

$$U_{\theta,i;j} = \begin{pmatrix} 0 & \cos(\theta) + i \sin(\theta) \\ \cos(\theta) - i \sin(\theta) & 0 \end{pmatrix}^{<i,j>}, \quad (22)$$

for any  $i \neq j$  and  $\theta \in \Theta$ .

Since  $\text{Tr}(ZU_{\theta,i;j}^* \mu U_{\theta,i;j}) = 0 \quad \forall \theta \in \Theta$  by Lemma 1, the coefficients of  $\sin 2\theta$  and  $\cos 2\theta$  in  $\text{Tr}(ZU_{\theta,i;j}^* \mu U_{\theta,i;j}) = 0$  will be zero. The coefficient of  $\sin 2\theta$  turns out to be  $\mu_{ij}(Z_{ij} + Z_{ji})$  and that of  $\cos 2\theta$  is  $i\mu_{ij}(Z_{ij} - Z_{ji})$ . It follows from (18) that  $Z_{ij} = Z_{ji} = 0$ .

Let us add to the set  $\mathbb{S}$  the matrices

$$V_{\theta,i;j} = \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ \sin(\theta) & -\cos(\theta) \end{pmatrix}^{<i,j>}, \quad (23)$$

for any  $i = 1, \dots, N-1, j = i+1$  and  $\theta \in \Theta$ . We use again  $\text{Tr}(ZV_{\theta,i;j}^* \mu V_{\theta,i;j}) = 0 \quad \forall \theta \in \Theta$  and identify the coefficients of  $\sin 2\theta$  and  $\cos 2\theta$  which are respectively  $\mu_{ij}(Z_{ii} - Z_{jj}) + \frac{\mu_{ii} - \mu_{jj}}{2}(Z_{ij} + Z_{ji})$  and  $-\mu_{ij}(Z_{ij} + Z_{ji}) + \frac{\mu_{ii} - \mu_{jj}}{2}(Z_{ii} - Z_{jj})$ . We obtain  $Z_{ii} = Z_{jj}$ . Since this is true for any  $i = 1, \dots, N-1$  from  $\text{Tr}(Z) = 0$  it follows that  $Z_{ii} = 0 \quad \forall i \leq N$ .

**Remark 3** *Although the hypothesis (18) may seem strong we believe that it is rather a technical requirement which could hopefully be relaxed in the future.*

## 5 Conclusion and discussion

Following previous works in [14] which study the properties of the control input-output map we investigate in this paper the singularity of the control to propagator map through the study of the linear independence of entries of the time-dependent coupling operator  $\hat{\mu}(t)$  (cf. (7)). We provide several sufficient conditions in terms of the evolution semigroup points that are reached during the propagation of Eq. (1). We expect that the criterion in (18) can be relaxed to accommodate even more general coupling operators.

## Acknowledgements

GT acknowledges support from the ANR-06-BLAN-0052 program and the MicMac project of INRIA Rocquencourt. TSH and HR were supported by U.S. Department of Energy.



## References

1. Albertini, F., D'Alessandro, D.: Notions of controllability for multilevel bilinear quantum mechanical systems. *IEEE Transactions on Automatic Control* **48**(8), 1399–1403 (2003)
2. Altafini, C.: Controllability of quantum mechanical systems by root space decomposition of  $su(N)$ . *J. Math. Phys.* **43**(5), 2051–2062 (2002)
3. Assion, A., Baumert, T., Bergt, M., Brixner, T., Kiefer, B., Seyfried, V., Strehle, M., Gerber, G.: Control of chemical reactions by feedback-optimized phase-shaped femtosecond laser pulses. *Science* **282**, 919–922 (1998)
4. Bardeen, C., Yakovlev, V.V., Wilson, K.R., Carpenter, S.D., Weber, P.M., Warren, W.S.: Feedback quantum control of molecular electronic population transfer. *Chem. Phys. Lett.* **280**, 151–158 (1997)
5. Bardeen, C.J., Yakovlev, V.V., Squier, J.A., Wilson, K.R.: Quantum control of population transfer in green fluorescent protein by using chirped femtosecond pulses. *J. Am. Chem. Soc.* **120**, 13,023–13,027 (1998)
6. D'Alessandro, D.: Introduction to quantum control and dynamics. Chapman & Hall/CRC Applied Mathematics and Nonlinear Science Series. Chapman & Hall/CRC, Boca Raton, FL (2008)
7. Girardeau, M.D., Schirmer, S.G., Leahy, J.V., Koch, R.M.: Kinematical bounds on optimization of observables for quantum systems. *Phys. Rev. A* **58**(4), 2684–2689 (1998). doi: 10.1103/PhysRevA.58.2684
8. Ho, T.S., Dominy, J., Rabitz, H.: Landscape of unitary transformations in controlled quantum dynamics. *Phys. Rev. A* **79**(1), 013,422 (2009). doi: 10.1103/PhysRevA.79.013422
9. Ho, T.S., Rabitz, H.: Why do effective quantum controls appear easy to find? *Journal of Photochemistry and Photobiology A: Chemistry* **180**(3), 226–240 (2006). URL <http://www.sciencedirect.com/science/article/B6TGY-4JRT33W-7/2/e487a7c6bfb086abc26371fbbe2d3e6c>
10. Hornung, T., Meier, R., Motzkus, M.: Optimal control of molecular states in a learning loop with a parameterization in frequency and time domain. *Chem. Phys. Lett.* **326**, 445–453 (2000)
11. Judson, R., Rabitz, H.: Teaching lasers to control molecules. *Phys. Rev. Lett* **68**, 1500 (1992)
12. Kunde, J., Baumann, B., Arlt, S., Morier-Genoud, F., Siegner, U., Keller, U.: Adaptive feedback control of ultrafast semiconductor nonlinearities. *Appl. Phys. Lett.* **77**, 924 (2000)
13. Levis, R.J., Menkir, G., Rabitz, H.: Selective bond dissociation and rearrangement with optimally tailored, strong-field laser pulses. *Science* **292**, 709–713 (2001)
14. Rabitz, H.A., Hsieh, M.M., Rosenthal, C.M.: Quantum Optimally Controlled Transition Landscapes. *Science* **303**(5666), 1998–2001 (2004). doi: 10.1126/science.1093649. URL <http://www.sciencemag.org/cgi/content/abstract/303/5666/1998>
15. Turinici, G., Rabitz, H.: Wavefunction controllability in quantum systems. *J. Phys.A.* **36**, 2565–2576 (2003)
16. Weinacht, T., Ahn, J., Bucksbaum, P.: Controlling the shape of a quantum wavefunction. *Nature* **397**, 233–235 (1999)
17. Wu, R., Dominy, J., Ho, T.S., Rabitz, H.: Singularities of Quantum Control Landscapes. *ArXiv quant-ph* p. 0907.2354 (2009). URL <http://arxiv.org/abs/0907.2354>